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SciVerse ScienceDirect
Physics Procedia 35 (2012) 145 – 150
Physics Procedia

Positron Studies of Defects 2011

Investigation of Radiation Affected High Temperature Superconductors - YBCO

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Abstract

In this paper, high temperature superconductors are studied in terms of radiation stability, which is necessary for application in fusion reactors. Perspective superconducting materials based on YBCO (Perkovskite structure) were measured by positron annihilation lifetime spectroscopy. Measurements were performed for samples prior to and after fast neutron irradiation in TRIGA MARK II reactor in Vienna. The samples demonstrated accumulation of Cu-O di-vacancies due to the irradiation. Nevertheless, the structure showed regeneration during thermal treatment by defects recombination. Positron spectroscopy results were complemented with values of critical temperature, which also showed changes of superconducting properties after the irradiation and the annealing.

Keywords: High Temperature Superconductor; Fast Neutron Irradiation; Critical Temperature; Cu-O Divacancies; Defect Concentration

1. Introduction

Recent research and development of fusion reactors are focused on materials resistance to high neutron and thermal loads. Although an emphasis is currently put on construction of ITER, there is also a progress in further concepts (DEMO, PROTO, etc.). High Temperature Superconductors (HTS) are perspective candidates for construction of magnetic systems in DEMO reactor [1]. Relative high critical temperature (T_c) of these materials enables a significant reduction of cooling costs. The most perspective HTS material in the view of recent research is $YBa_2Cu_3O_{7-\delta}$ (Perovskite structure) with the critical temperature about 92 K. This material is specifically interesting because of its high critical current densities at high fields and elevated temperatures.

In fusion facilities like DEMO, fast neutrons (up to ~ 14MeV) produced during nuclear reactions can form defects in materials structure due to atom knocking-out. The most radiation loaded materials of the fusion reactor are situated according to [2] in the inner wall – within inboard blanket, shielding as well as superconductors affected by

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 doi:10.1016/j.phpro.2012.06.026

fast neutron flux density ~ 10^{14} m^{-2} s^{-1} (See Fig. 1). Neutron fluence estimated during projected lifetime is close to $2.5x10^{22}$ $m^{-2}MW^{-1}$ [3].

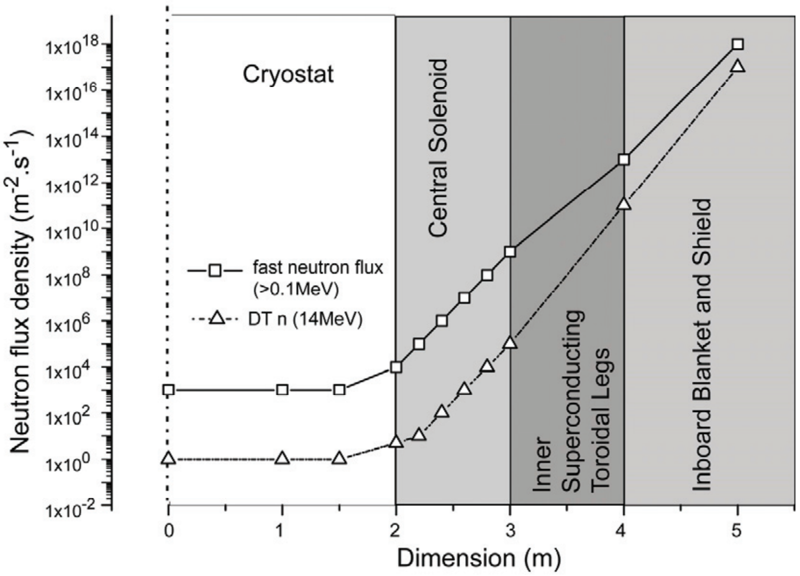

Fig. 1 Estimated neutron flux within inboard part of the fusion reactor DEMO.

Superconducting properties can be improved or optimized by applying of flux pinning [4], which is an important phenomenon in all industrial superconductors. Defects as miss-oriented grains, twin boundaries, dislocations, voids and precipitations are responsible for high currents transportation [5]. Therefore defects introduced by fast neutron irradiation can be partly effective pinning sites [6, 7] and can significantly improve the superconducting properties of $YBa_2Cu_3O_{7-\delta}$ [8]. Nevertheless, microstructure is being damaged and mechanical properties are reduced during this improvement.

In this paper, the accumulation of small defects is investigated by positron annihilation lifetime spectroscopy (PALS) and influence of the small defects on the critical temperature is also observed. The samples were irradiated in an experimental reactor by fast neutron fluencies up to 6.10^{21} m^{-2}.

2. Experiment

Two compositions of $YBa_2Cu_3O_{7-\delta}$ (Perovskite structure) were studied in terms of radiation stability after irradiation:

i) YBCO – sintered material without texture, density of the bulk reached 4.235 g/cm^3.

ii) MS2F – multi seed bulk manufactured by "melt texture growth" process [9]. MS2F is composed of 2 different yttrium structures ~70% Y123 and ~30% Y211 (Y_2BaCuO_5) with additional element platinum Pt (0.1%). Its density is 5.274 g/cm^3.

Although a superconducting magnetic system in the fusion reactors will be constructed from coated conductors, the experimental techniques applied in this paper required larger diameter of samples. Therefore, the bulk materials were investigated.

The investigated samples were irradiated in the experimental TRIGA MARK II reactor in Vienna. A reactor power was 250 kW with thermal/fast neutron flux density of 4.2/5.3 x 10^{16} $m^{-2}s^{-1}$. A first irradiation level corresponds to a fast neutron fluence of 1.2 x 10^{21} m^{-2}. A second irradiation level achieved a fluence of 6 x 10^{21} m^{-2}.

After the measurement of irradiated samples, they were annealed at 250°C for 4 hours in air for purpose of observation of defect recombination and structure regeneration.

The experimental measurement was performed by using of positron annihilation lifetime spectroscopy (PALS) [10]. Defect size and defect concentration were estimated in both samples prior to and after the experimental treatment. Table 1 shows values of positron lifetime characteristic for $YBA_2Cu_3O_7$ bulk together with values for common defects within this structure.

Table 1. Typical lifetime for bulk and defects [11-13].

Structure/ Defect	Lifetime (ps)
YBa2Cu3O7 (Orthorombical structure)	193
O mono-vacancy	~ 170
O vacancy cluster (2 -4 vacancies)	181 - 190
Cu(1) mono-vacancy	207
Cu (2) mono-vacancy	182
Y mono-vacancy	206
Ba mono-vacancy	263
Cu (1) - O (1) di-vacancy	236

PALS results can be used for identification of defects and quantification of their concentration. There is an assumption that only one type of defects will be in predominance (smaller defects). The bigger defects (e.g. voids, vacancy clusters, if exist) will be included in the lifetime of positronium, which is described with very small intensity. If we neglect existence of bigger defects and only focus on smaller defects only, calculation of their concentration can be performed according to [14 - 16].

Measurement of critical temperatures was performed in 1 T *Superconducting Quantum Interference Device* (SQUID) magnetometer [17]. The transition temperature of a superconductor is defined as the temperature below which a superconductor exhibits zero resistance and perfect diamagnetism in the limit of zero applied magnetic field and current.

3. Results and Discussion

Data obtained by PALS measurements were separated into three components by code Lifetime 9 [18] according to the Standard trapping model. The value of FWHM parameter describing time resolution of measuring equipment [19] was close to 220ps. Fit Variant (reduction of chi-square) achieved value in range (1; 1.1), which means that the goodness of the fit was sufficient and the aberration of the fit was below 0.1%.

The shortest lifetime (LT1) achieved values up to 190ps (see Tab. 1), which describes the positron annihilation in defect-free structure reduced due to positron trapping in defects. The LT1 was significantly reduced mostly in irradiated samples. The second positron lifetime (LT2) within the range 220 and 600ps characterizes vacancy type defects and microvoids. The last lifetime (the longest one) is not further mentioned, because it only corresponds to in-flight or surface annihilation.

The positron lifetime (LT2) for the non-irradiated samples of $YBa_2Cu_3O_{7-\delta}$ achieved values 310ps (YBCO) and 373ps (MS2F). The intensity of this positron lifetime (I2) is up to 10%, i.e. MS2F (10.5 %) and YBCO (4 %) (See Fig. 2).

After the irradiation up to 1.2 x 10^{21} m^{-2}, the intensity I2 grew about 50% for the both materials, i.e. MS2F (ΔI2=58.1%) and YBCO (ΔI2=60.9%), which demonstrates defect accumulation into the samples during material exposition in the reactor. The sample of MS2F with the LT2 ~ 250ps probably contains the same type of defects as YBCO (232ps), but with lower defect concentration related to intensity difference of 2.7%.

The further irradiation (up to 6 x 10^{21} m^{-2}) caused accumulation of new defects and the defect concentration increased again. The LT2 was still around 240ps which indicates accumulation of the same defects like during the first irradiation. The change of defect concentration was significant for both materials, MS2F - ΔI2=19.2% and YBCO - ΔI2=17.1%.

Results for annealed samples demonstrated decrease of the defect concentration. The small defects created during irradiation were able to recombine. Therefore the superconducting properties of the structure should be also regenerated and similar with properties of the non-irradiated samples. However, the effect of recombination of smaller defects is not the same as for bigger ones. The lifetime describing the bigger defects grew which demonstrates significant accumulation of these defects during the irradiation. The bigger defects were not able to recombine during the annealing; therefore they remain in the structure.

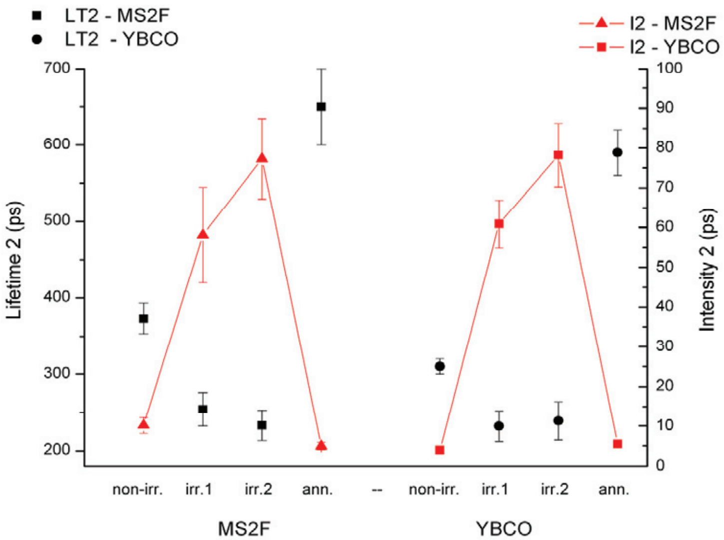

Fig. 2 PAS results for non-irradiated samples of YBCO and MS2F as well as for samples in both irradiated levels and after annealing.

Measured lifetime (LT2) for the non-irradiated sample and also for the annealed samples indicates containing of big vacancy clusters (~ 350ps [20]) or microvoids (~ 600ps corresponds to 50 vacancy cluster [16]), which were expected on the ground of already published data. In irradiated samples, di-vacancies probably started to dominate; although the bigger defects are still located there.

Defect concentration of found Cu-O di-vacancies was quantified according to equations published in [14 - 16]. Calculation was performed only for the irradiated samples where the bigger defects were neglected and only di-vacancies were observed. Results (see Table 2) show visible increase of the defect concentration after the irradiation for both materials YBCO and MS2F as was assumed. This calculation corresponds to changes of observed mean lifetimes (MLT).

Table 2. Calculation of the defect (Cu-O divacancy) concentration in samples.

Sample		Defect concentration c_D (ppm/at.)
MS2F	Non-irradiated (microvoids)	-
	1st irradiation (Cu-O di-vacancies)	1.50
	2nd irradiation (Cu-O di-vacancies)	3.75
	Annealed (microvoids)	-
YBCO	Non-irradiated (microvoids)	-
	1st irradiation (Cu-O di-vacancies)	1.68
	2nd irradiation (Cu-O di-vacancies)	3.96
	Annealed (microvoids)	-

The MLT increased much more for irradiated YBCO (1st irradiation ΔMLT = 10 ps, 2nd irradiation ΔMLT = 18 ps) than for MS2F (9 ps and 3 ps). The measurement uncertainty is less than 2 ps. Therefore these results indicate significant change in the defects concentration after the irradiation, mainly in YBCO; although MLT of MS2F is always higher. MS2F contains much more defects in total.

After the annealing, MLT of both samples decreased similar. YBCO achieved a little bit higher MLT than for the non-irradiated sample, which means that the structure of YBCO was not fully regenerated. On the contrary, MLT of annealed MS2F is smaller than for the basic sample without the experimental treatment. This structure shows almost complete regeneration of microstructure in terms of positron data.

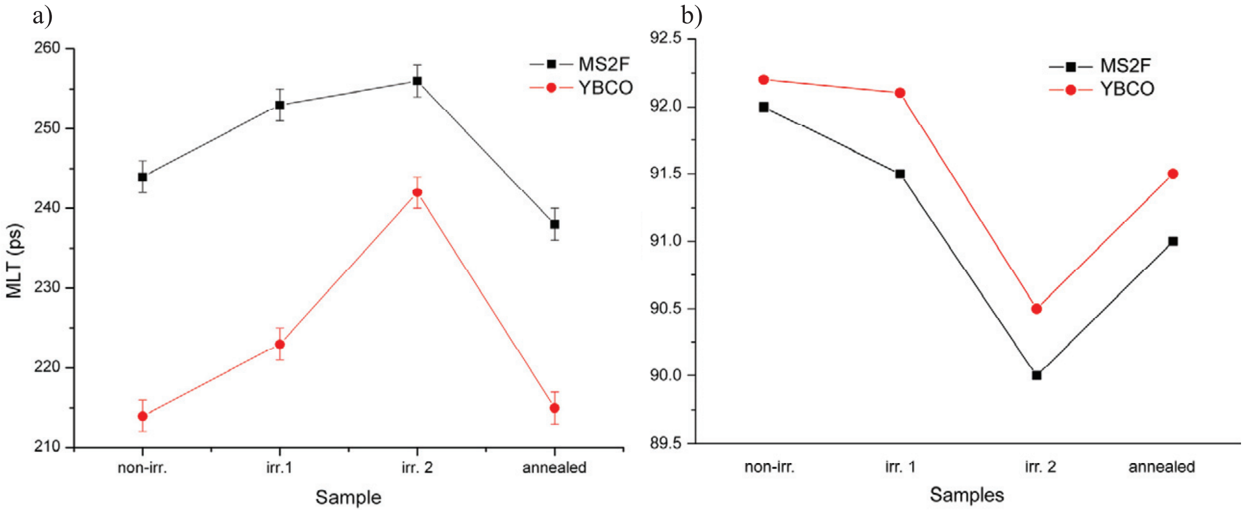

Fig. 3 The change of positron mean lifetime (MLT) after irradiation and annealing (a); Critical temperatures measured in SQUID magnetometer (b).

The critical temperature also indicated changes for both samples (See Fig. 3). After the irradiation as well as the annealing, the critical temperature was always higher for YBCO, although difference between YBCO and MS2F was small for each measurement.

The results after the irradiation showed undesirable phenomena of T_c reduction for both samples. The changes of the critical temperature after the irradiation were less significant for the YBCO sample. This result also indicates that the superconducting properties (the critical temperature) were worsened after the irradiation and the assumption about formation of new effective flux pinning sites was not confirmed.

4. Conclusion

High temperature superconductors are materials with high potential for various applications and can effectively replace conventional superconductors in fusion facilities. $YBa_2Cu_3O_{7-\delta}$ is one of the most perspective materials in the view of its high critical temperature. Recent research proved necessity an appropriate structure with demand on larger defect concentration. This is not typical for another material, where defects cause degradation of structure and elimination of mechanical properties. Nevertheless, high defect concentration in superconductors can support superconducting properties. The actual study of influence of neutron flux and gamma radiation indicates visible change of superconducting properties of $YBa_2Cu_3O_{7-\delta}$ after the irradiation. The critical temperature decreased for both investigated samples – pure YBCO and multi-seed MS2F (with addition of platinum).

The non-irradiated samples contain low concentration of big defects and presence of small defects was not proven. After the irradiation, mainly smaller defects Cu-O di-vacancies accumulated into the structure. Defects formed due to neutron irradiation dominate with their quantity over defects located in the samples before the radiation treatment.

The annealing of samples caused higher mobility of the small defects and their merging to the bigger ones or their recombination. The $YBa_2Cu_3O_{7-\delta}$ lattice is partially regenerated by this process which is evident from the critical temperature enhancement.

Acknowledgement

N. Hari Babu (University of Cambridge) and P. Diko (Slovak Academy of Science) are acknowledged for preparation of investigated materials. Further, M. Eisterer and H.W. Weber (Vienna University of Technology) are also acknowledged for performance of the critical temperature measurements and samples irradiation. This study was supported by grants VEGA 01/0129/09 and the Nano Engineered Superconductors for Power Applications (NESPA).

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